

MECHANICAL PROPERTIES OF FIBRE/ FILLER BASED POLY(LACTIC ACID) (PLA) COMPOSITES : A BRIEF REVIEW

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Abstract

Being a biodegradable polymer, poly(lactic acid) (PLA) based composites receive greater preference over non-biodegradable plastics. Poly(lactic acid) has to find its place in various applications such as polymer composites, agriculture, biomedical, etc. Polymer composites based on PLA possess comparable mechanical strength, endurance, flexibility and endures future opportunities. Several combinations of natural fibers and filler-based PLA composites have been fabricated and investigated for physical and mechanical changes. Moreover, several biopolymers and compatibilizers are added to PLA to provide rigidity. The paper presents a tabulated review of the various natural fiber/filter-based PLA composites and the preparation and outcomes. In addition, enhancement made by the reinforcement of nano filler in the PLA are also discussed in brief. The significance of PLA in the biomedical application has been discussed in brief. The paper also shed lights in the social and economic aspects of PLA.

Keywords

poly(lactic acid) (PLA); biodegradable composites; mechanical properties.

Introduction

The increase in carbon emission and the excess usage of plastic polluting the air, land, and water are the major causes for the substitution of plastic for biodegradable polymers [1–3]. In fact, biodegradable polymers have become necessary to maintain an ecological balance between the source and waste production, ensuring minimum waste accumulation in the environment [4–6]. Millions of plastic commodities are being manufactured in everyday life, and almost equal is disposed of in the environment. These plastics are reinforced with various types of fibers and fillers to improve mechanical strength [7–9]. The most typical kinds of plastics used in daily practice are polypropylene, polyethylene, polystyrene, polyvinyl chloride, and polyurethane. The polymers mentioned above take a very long time to degrade hence called plastics or non-biodegradable polymers, due to the presence of a long polymeric chain and shows detrimental effects to the fertility of soil when dumped [10]. In an intention to resolve the issue of biodegradability, various natural and synthetic biopolymers were introduced in the market. Among them, PLA, a synthetic biopolymer, was found suitable to substitute the existing polymers due to good compatibility with other biopolymers to form blends, comparable tensile and flexural strength, large deformation, and remains thermally stable at elevated temperature [11]. PLA can be easily

manufactured by the process of fermentation and consume less energy during processing. It is cheap and available on the market. The alarming situation of CO₂ emission by the use of non-biodegradable polymers can be curbed down by using PLA. Problem of disposing plastic in the landfills and aquatic can also be controlled to some extent. Using PLA as a bio material is not only beneficial economically but also favourable for environment and social ecosystem.

Many biopolymers are used in biocomposites with projected applications in sectors like automobile, marine engineering, aerospace, electronics, and household ware [12]. In the last decade or two, poly(lactic acid) has been extensively used as a matrix material for the fabrication of bio-composites with several types of reinforcements such as fiber (natural or synthetic), fillers (bio filler or nanofiller), metallic, and ceramics [13]. The properties of PLA-based composite fabricated in the past differ in method of fabrication, weightage of reinforcement, addition of compatibilizers etc. which will be discussed in the literature section.

Process of manufacturing PLA based composites

Composites based on PLA are prepared by various processes depending upon the nature of reinforcement and type of composite to be prepared. For instance, if the reinforcement is filler type, then twin-screw extruder (Figure 1) and injection moulding is used [14]. Various natural fillers such as wood flour, silicates, carbonates are reinforced in this process. In the case of fiber mat, a compression moulding machine is employed. Several kinds of natural and synthetic fiber can be reinforced with PLA in compression moulding machines such as hemp, kenaf, sisal, coir, wood, carbon, etc. [15–17]. Both the processes are equipped with the provision of a heating element to melt the pellets of PLA. Usually, the temperature range of heating elements is kept between 170-185° C. Blends of PLA have been often prepared to provide rigidity in which various kinds of biopolymers are added like polyethylene glycol (PEG), poly(butylene succinate) (PBS), poly(hydroxy acid) (PHA), natural rubber (NR), polycaprolactone (PCL) and poly(hydroxy butyrate) (PHB). These blends are prepared by a melt blending process in which polymers are mixed in a beaker and stirred [18]. Thereafter, the required reinforcement is added to the blend. Injection moulding or melt screw moulding is advantageous in several ways over other manufacturing processes like compression moulding, hand lay-up, vacuum assisted resin transfer moulding etc. for example low running cost, specimen for specific dimension can be made directly by using die at the exit, complex material can be fabricated easily, variety of polymer can be easily processed due to the presence of heating element, higher rate of production, longer working life because of low applied pressure and cleanliness of fabricated material due to lower possibility of contamination.

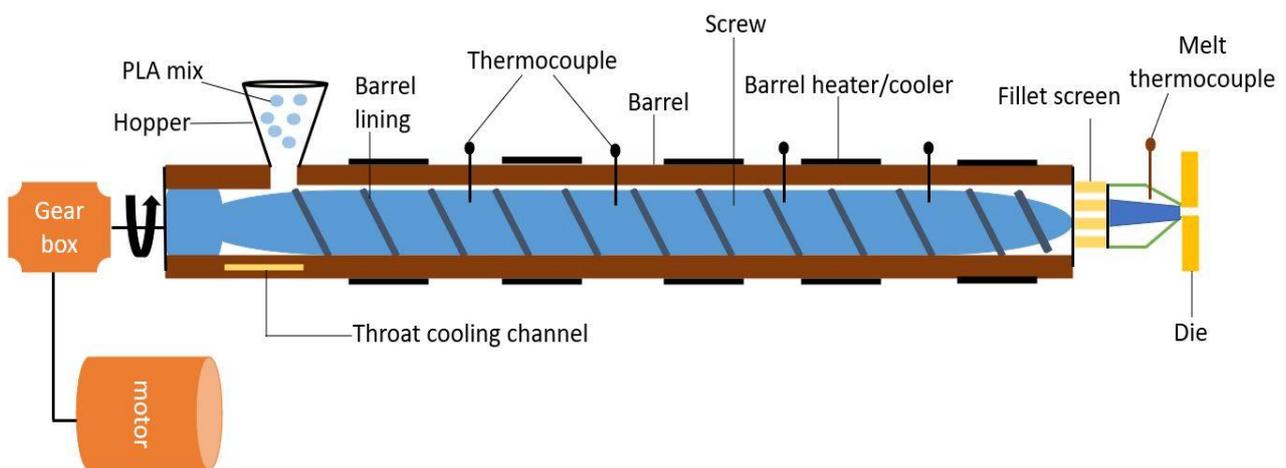


Figure 1. Melt extruder for the fabrication of composite based on PLA [14].

Work done on Poly(lactic acid) based composites:

Table.1. PLA based biopolymer reinforced with natural fibre.

S. No.	Material	Process parameters	Particulars of research	Ref.
1	PLA grafted with 3 % Maleic anhydride reinforced with 30 % and 50 % Wood flour	Twin-screw corotating extruder followed by injection moulding.	Reduced flexural modulus, storage modulus, and tensile modulus were observed whereas failure strain, coefficient of thermal expansion, and melt flow index improved. Thermal stability was enhanced while the crystallinity of PLA decreased.	[19]
2	PLA with 5 % to 20 % fleece fiber and coir fiber	Hot-press moulding at a temperature of 170° C, the pressure of 10MPa, and moulding time of 10 minutes.	An increase in tensile strength of the composite was observed with an increase in the percentage of fleece fiber but it decreases as the fleece fiber weightage increases to 20 %. Modulus of elasticity increases with an increase in the percentage of fleece fiber.	[20]
3	PHB and PLA with 30 % short and long pulp fiber.	Fibers were mixed with a blend of PHB and PLA in a torque rheometer at 190 °C	The impact strength was enhanced by a factor of 1.3, while tensile strength was enhanced by a factor of 1.5 as compared to the virgin blend of PLA and PHB.	[21]
4	PLA with olive husk flour at 10, 20, and 30 % reinforcement	Composites were prepared with the extrusion-injection moulding method.	Young modulus increased by almost 27 % from the initial value with the addition of olive husk flour. However, the tensile strength and elongation at break were reduced owing to poor dispersion and weak interfacial bonding. Treatment of olive husk flour resulted in an enhancement in interfacial bonding and improved mechanical and thermal properties.	[22]
5	PLA and short kenaf fiber	Blend of PLA, and short kenaf fiber was prepared by melt blending in Brabender internal mixer at 60 rpm and 170° C for 10 minutes.	The addition of short kenaf fiber improved the tensile strength and tensile modulus of the composite up to 30 wt. % of the reinforcement. At 40 wt. % and 50 wt. % of kenaf fiber reinforcement, the PLA matrix unable to wet the fiber and resulted in reduced tensile strength and tensile modulus. Change in the crystallinity and porosity of PLA was also observed due to the reinforcement of short kenaf fiber	[23]
6	PLA, PBS, PCL, PBAT, NR, PCL and grass fiber	Blending of polymer was carried out in laboratory and reinforced with grass fiber via injection moulding	Blending of polymer with NR significantly improved the impact strength. Improvement in the tensile strength and tensile modulus with the incorporation of grass fiber was observed but the impact strength and elongation at break reduced.	[24]

7	PLA and Basalt fiber	Mixture of PLA and dried basalt fiber were prepared in twin screw extruder and then passes through injection moulding for the preparation of composite samples	The mechanical properties of PLA increased linearly with the increase in the percentage of basalt fiber in composite. The highest improvement in the mechanical properties were obtained at 30 % weightage of basalt fiber.	[25]
8	PLA and continuous carbon fiber	Fused deposition modelling	The carbon fiber reinforced PLA composite yielded enhanced tensile and flexural strength.	[26]
9	PLA, chitosan and carbon fiber	Solvent blending and immersed glow moulding process	The presence of chitosan in carbon fiber PLA composite helped bridge the gap between the fiber and matrix, resulting in improved shear and bending modulus. The rate of bio degradation of the composite accelerated with the addition of chitosan.	[27]
10	PLA and bamboo fiber	Composites were prepared in injection moulding machines	Bamboo fiber lowered the glass transition temperature, storage modulus and loss modulus of the PLA. Moreover, the mechanical properties of PLA significantly reduced.	[28]
11	PLA, benzoyl peroxide, banana fiber and sisal fiber	Composites were prepared by twin screw extrusion followed by injection moulding process.	Excellent improvement in the mechanical properties of PLA especially in tensile and flexural strength attributed to the treatment of fibers with benzoyl peroxide.	[29]
12	PLA, sugar beet pulp and sorbitol and glycerol as plasticizer	Composites were prepared by twin screw extrusion followed by injection moulding process.	Tensile strength reduced by 25 % with the reinforcement of 30 % of sugar beet pulp whereas the elongation at break increases. Specific mechanical energy of PLA reduced from 1400 J/gram to 1000 J/gram.	[30]
13	PLA and okra fiber	Co-rotating twin screw micro extruder	The stiffness of PLA increased with the addition of okra fiber. The morphological study revealed that fiber diameter was reduced by the chemical treatment but the interlocking enhanced between the fiber and matrix.	[31]
14	PLA and chitin	Melt blending was used to prepare the blend of PLA and chitin	The drop in peak degradation temperature of PLA was observed with the addition of chitin. Char formation increased as PLA became more hydrophobic leading to enhancement in tensile and flexural strength.	[32]
15	PLA, carbon fiber and SEBS-g-MA as compatibilizer	Composites samples were prepared by single screw extruder	Tensile strength, tensile modulus and flexural modulus decreased whereas flexural strength increased marginally and significant improvement was observed in the impact strength of PLA with the addition of chitin.	[33]

16	PLA, cassava flour, pine apple flour and ash.	Samples were prepared by injection moulding machine	Mechanical properties of PLA enhanced up to 30 % of both cassava and pine flour reinforcement but started decreasing when the reinforcement weightage increased to 40 %.	[34]
17	PLA, basalt fiber and wood flour	Co rotating twin screw extruder was used to fabricated the composite samples	SEM micro graphs confirm the homogenous mixing of reinforcements in PLA. There was no improvement observed in the mechanical properties of PLA with the incorporation of basalt fiber rather decreased with the incorporation of wood flour in PLA.	[35]
18	PLA, epoxidized natural rubber and lignocellulose fiber	Composites samples were prepared by using a vulcanized mould in press moulding	On mixing natural rubber with PLA, the elasticity improved. The glass transition temperature of PLA increased with the addition of lignocellulose fiber. It was also found that the decomposition rate of PLA accelerated due to the incorporation of silane-treated lignocellulose fiber.	[36]
19	PLA and coir fiber	Samples were prepared with co-rotating twin screw extruder	At 30 % reinforcement of coir fiber the tensile strength reduced to 52.8 MPa from 57.4 and elongation of break reduced to 2 % from 3.8 %. The tensile modulus was found to be increased from 4 GPa to 4.8 GPa.	[37]
20	PLA and hemp fiber	Composite samples were prepared by twin-screw extrusion and injection moulding	At 40 wt. % loading of hemp fiber in PLA, the flexural strength, flexural modulus, and impact strength were found to be maximum with an increment of 62 %, 90 %, and 68 % respectively as compared to virgin PLA.	[38]
21	PLA, 25 wt. % cotton linters and 50 wt. % Maple fiber	Composites samples were prepared via compounding followed by injection moulding	Marginal increase in the overall density was achieved with the addition of 25 % cotton linters in the PLA. Flexural modulus of PLA was increased by 56 % and 123 % by the addition of cotton linters and Maple fiber respectively. Significant improvement in the impact strength of PLA was also observed.	[39]
22	PLA, office waste paper and PBAT	Composites were prepared by melt blending and injection moulding	The impact strength of PLA was enhanced by 291 % at 20 wt. % of office waste paper but the flexural modulus and flexural strength decreased. PBAT addition in PLA increases the water uptake stepwise.	[40]
23	PLA, kenaf fiber and rice husk	Compounding of PLA with fibers were carried out followed by samples preparation by injection moulding	The flexural modulus of PLA increased around 32 % and 18 % with the addition of kenaf fiber and rice husk respectively. However, flexural strength and impact strength decreased.	[41]
24	PLA, bamboo fiber and silane coupling agent	Composite samples were prepared by injection moulding	Storage modulus and loss modulus of PLA decreased as the weight percentage of bamboo fiber increased in the composite.	[28]

25	PLA, benzylated treated rice straw and nano clay	Solvent casting method was used to prepare the composite samples	Reduction in the peak intensity of the cellulosic hydroxyl group was observed with the addition of treated rice straw due to the formation of aromatic bonds. A slight increase in the crystallinity of PLA was observed due to strong molecular interaction between fiber and PLA.	[42]
26	PLA and rice straw	Alkali pulping and benzylation was performed on the rice straw followed by twin screw extruder for the composite fabrication	The addition of rice straw in PLA resulted in the lowering of glass transition temperature and melting temperature. Early degradation of the composite was confirmed by the SEM micrographs.	[43]
27	PLA and thin strip of bamboo	Strips of bamboo were bonded in PLA with the help of compression moulding machine for the fabrication of composites	Excellent enhancement in the mechanical properties of PLA was obtained due to the influence of the node. Highly stiffed composite can be fabricated via compression moulding process.	[44]
28	PLA and 10 wt. % PEG	Melt blending and co rotating twin screw extruder	PEG plasticized the PLA but the mechanical properties were reduced.	[45]
29	PLA, raw fiber obtained from acid-catalysed hydrolysis and delignified fiber	Samples were prepared by co rotating twin extruder	Marginal improvement was observed in the tensile strength of PLA with the addition of raw fiber. Formation of pores and voids took place due to the addition of oat delignified fiber.	[46]
30	PLA and nanometric and micrometric cellulose filler	Solvent casting method was used for the fabrication of blend	Higher young modulus and tensile strength were obtained for nanometric filled PLA composite as compared to micrometric filled PLA composite	[47]
31	PLA, starch, cellulose fiber in powder form and carnauba wax,	Foam composite was prepared by compression moulding	The foam containing 9 % PLA showed the highest improvement in tensile strength with an increment of 31 % as compared to foam without PLA.	[48]
32	PLA with 10 % copper fibers	Compression moulding technique	Viscoelastic behaviour of PLA enhanced with the addition of copper fiber. The PLA became more thermally stable due to the reinforcement of copper fibers.	[49]

The results reported in the tabulated form (Table 1) conclude that reinforcing PLA with fiber/filler reduces the strength but enhances the modulus and stiffness. Various compatibilizers are added in the PLA to improve the mechanical interaction between fiber and PLA and enhances the melting point temperature. The addition of copolymer increases toughness and crystallinity of PLA due to formation of cross linkage. In addition, the dynamic mechanical properties such as loss modulus and storage modulus are also enhanced by the addition of copolymers.

Poly(lactic acid) based nano composite

Various inorganic and organic materials can be incorporated in PLA to enhance its mechanical and thermal properties. These incorporations, when performed at nanoscale then it is called nano composites.

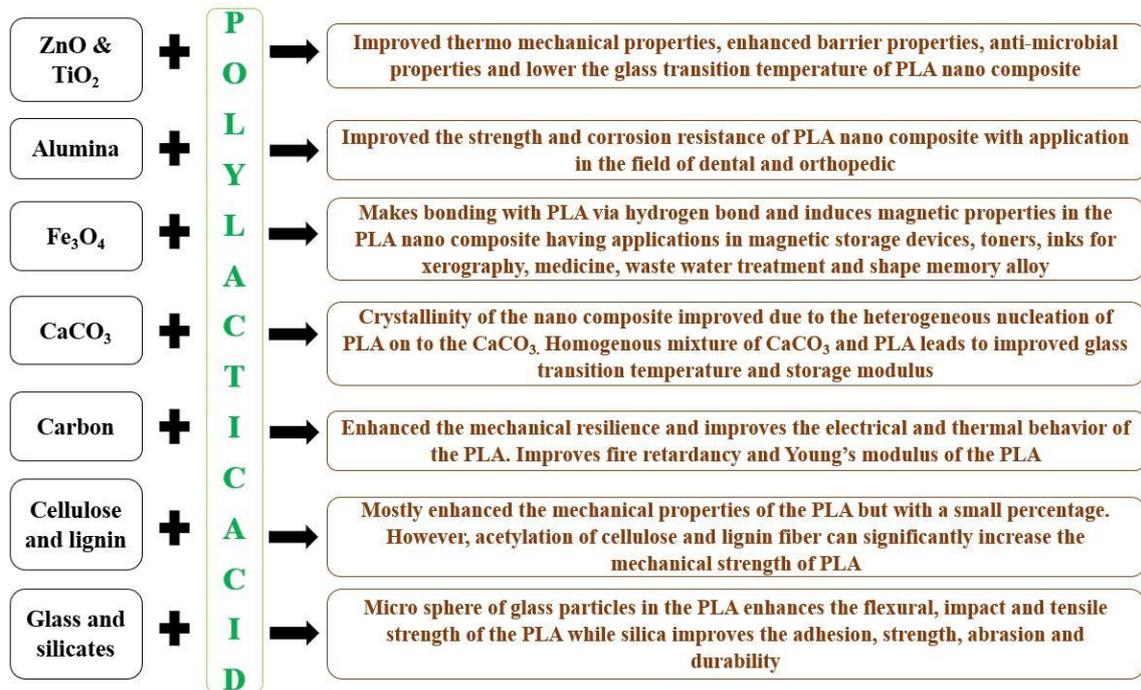


Figure 2. PLA based nano composites [50–70].

A minimal amount of nanofiller is sufficient to improve the mechanical and thermal properties of the PLA significantly. Due to the high surface to volume ratio of nano filler, it is usually reinforced in a small amount i.e. upto 5 %. It generally occupies the vacant position in the matrix and provides rigidity to the composites leading to high impact and tensile strength. Significant enhancement in the strength of PLA has been reported in the past by reinforcing various nano fillers such as ZnO, silicates, CaCO₃, Fe₃O₄, TiO₂ etc. nano fillers not only provides strength to PLA but also improves thermal characteristics. Various nano fillers-PLA composite and their properties enhancement is shown in Figure 2.

Application of PLA based composites

Despite being subjected to various composite manufacturing as a matrix material, poly(lactic acid) has also been used in the field of biomedical. It has favorable characteristics such as biodegradability, biocompatibility, hydrolyzed under environmental conditions, low immunogenicity, etc. that makes it a suitable polymer for various biomedical applications, as illustrated in Figure 3. It is used in orthopedic implants in which biocomposites based on PLA are used for bone fixation and implant for tissue recovery. Blends of PLA, polyglycolide (PGA), and di-oxanone are used for orthognathic surgery, a pretreatment for maxillofacial fractures. PLA blends provide comfort and relief to the implants and eliminate the secondary process of healing [71]. Several drug delivery systems are prepared by PLA. A combination of lactide group and glycolic acid are used for lipophilic drugs, chemotherapeutic drugs, and therapeutic agents. Antigen release can also be accomplished by combining microsphere of chitosan with poly(glycolic acid) [72].

Scaffolds prepared from PLA and collagen are effectively used to stimulate and proliferate cell generation in tissue engineering. The augmentation of soft tissue damage has been successfully achieved by phosphate-PLA biocomposite for bone tissue engineering. Successfully implementing composite based on poly(lactic acid) as an antimicrobial agent has been accomplished for bidding micro-organism growth in water sanitation plant [73]. Research and development are also in progress to develop stents based on PLA used for the clinical problem that arises in the ureteral system. Various electronic controller systems are now being fabricated from poly(lactic acid) leading to mutual compatibility, less space, efficient power, and reliability as compared to conventional material. Teabags, packaging materials, and other household ware are also manufactured with poly(lactic acid).

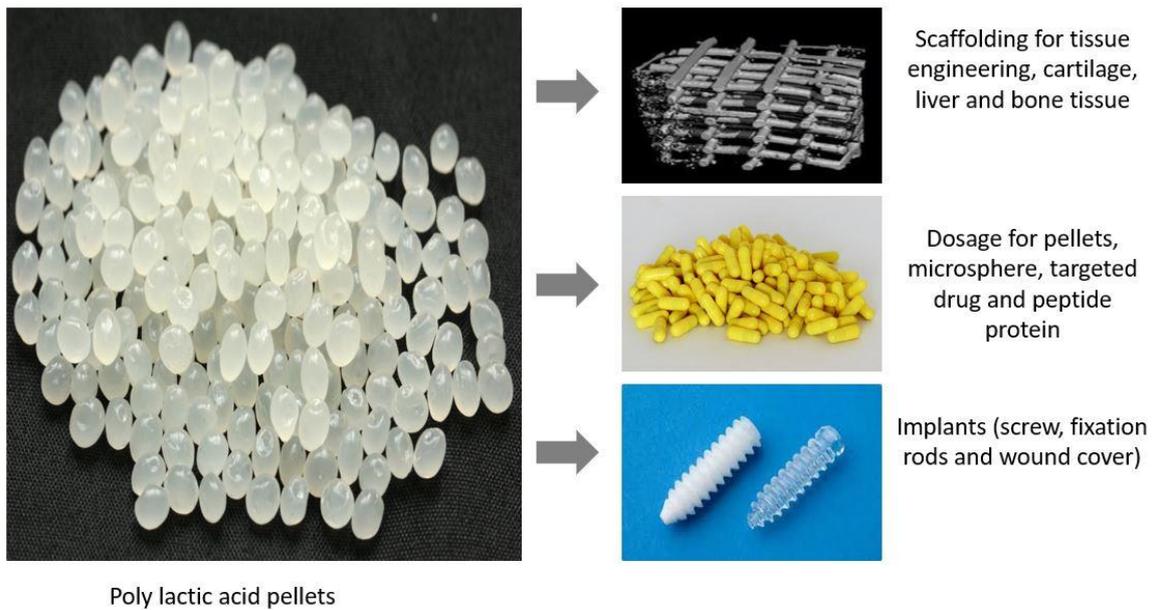


Figure 3. Biomedical scope of poly(lactic acid) [71].

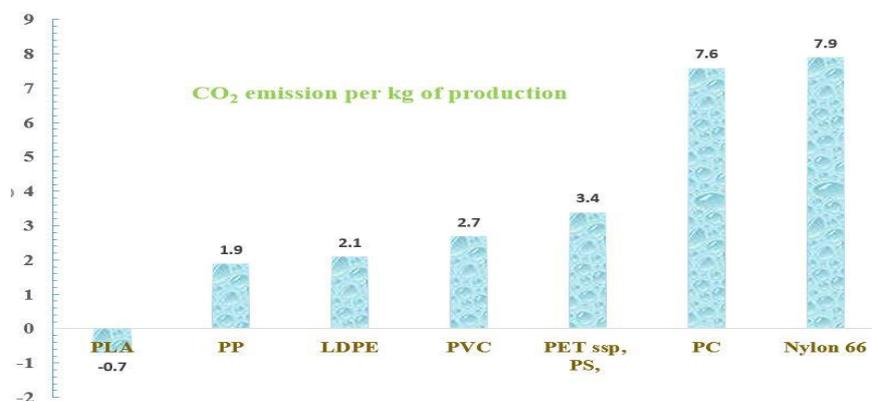
Impact

Cost of production of any material is the crucial parameter which decides its viability and future sustainability in the industry. Today, various companies like NatureWorks, Futerro, Corbion etc. are providing PLA at reasonable cost. The quality of PLA offered by these companies is worth buying but the original production cost is unknown to almost everyone except few producers. To assess the original production cost of the PLA a number of experiments were performed. Raw material used for the production of PLA were potato starch, wood extract, cassava extract, food waste, triticale and corn. The cost of production incurred in the analysis varied according to the method adopted. The detailed cost analysis of PLA production is explained in Table 2. The variation in the cost is the result of method used in the production, type of feed stock and other direct and indirect cost considered in the investigation therefore a comparison among the studies on the basis of production cost will not be justified. Nevertheless, various methods of producing PLA at different working conditions have evolved through research and technical advancement.

By using different types of feedstock and technique, we can lower down the production cost of PLA as shown in Table 2. However, the commercialization of the method will depend on the balance between its cost and environmental effect. PLA is well-known for its low CO₂ emission and the feedstock i.e. corn, consumes CO₂ during its cultivation [74]. Comparing the carbon emission of PLA production with other typical polymers, it was found that PLA has negative greenhouse gas emission [75]. This makes PLA act as a greenhouse gas sink and coupling this with latest process technology will definitely lead to the formation of a green environment.

Table 2. Cost analysis of PLA produced by different method [76–79].

Feedstock	Technique	Cost incurred	Remark
Potato starch	Bacterial fermentation (Green field technology)	Cost of dextrose, waste disposal cost, revenue cost, fixed cost such as electricity cost and overhead cost	\$ 2.9 US production cost per kg of PLA
Sugar extract from wood	Bacterial fermentation (Brownfield technology)	Lowers the capital cost, annual operating costs, including taxes, insurance, maintenance, return on investment and other expenses	\$ 2.62 US production cost per kg of PLA
Corn starch	Yeast fermentation	Lower the waste cost and process cost	\$ 2.12 US production cost per kg of PLA
Cassava root	Cargill dow process	Costs of PLA production, raw material costs (cassava roots and chemicals), capital costs, labour costs, operating costs, and waste treatment costs, environmental cost and cost of CO ₂ emissions resulting from electricity and fuel consumption	\$ 2.71 US production cost per kg of PLA
Cassava starch	Cargill dow process	Raw material costs (cassava starch, capital costs, labour costs, operating costs, and waste treatment costs, environmental cost and cost of CO ₂ emissions and waste cost (gypsum)	\$ 2.82 US production cost per kg of PLA
Cereal crops (triticale)	Baseline technology	Costs of raw materials (biomass and chemicals), energy and operating materials, maintenance and repair, labour, operating materials, insurance and overhead, administration, distribution and sales	\$ 1.2 US production cost per kg of PLA
Corn grains	Bio refinery	Equipment cost, raw materials cost, energy cost, labour cost and costs of producing lactic acid from bacteria, fungi and yeast-based fermentation	\$ 0.84 US production cost per kg of PLA

Figure 4. CO₂ emission by various polymers [80].

Conclusions

The fabrication of the biocomposites in the coming future will be given preference over non-biodegradable composites due to the strict regulations as specified by the United Nation environmental program (UNEP) for the prevention of the environment from the hazardous effect of the polymer waste where poly(lactic acid) can play a crucial role. Different types of biopolymers compatible with PLA need to be exposed to expand its scope for the fabrication of sustainable material that could perform in real-time conditions.

Poly(lactic acid) is readily available in pallet form and is widely used for the manufacturing of bio-composites particularly via melt extruder and injection moulding. Various favourable changes in mechanical and wear characteristics of PLA have been reported along with some future suggestions. PLA showed decreasing characteristics of tensile and flexural strength with the addition of filler which can be modified with the use of compatibilizers or mixing of suitable biopolymers.

Biodegradable implants are primarily designed from PLA-based composites and are expected to be made from pure PLA in the near future also. The emerging biomedical tailorable applications will be made from the PLA and its copolymer blends such as glycolic acid, caprolactone, and polyethylene glycol.

Conflict of interest

There are no conflicts to declare.

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